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REMOVAL OF RADON FROM INDOOR AIR BY ACTIVATED CARBON AND SOLID DESICCANTS

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ABSTRACT

The adsorption capacity of a coal-based activated carbon for radon from dry nitrogen is significantly higher than that of either silica gel or molecular sieves, although the capacity of activated carbon decreases sharply in the presence of water vapor. However, solid desiccant systems employing either silica gel or molecular sieves can be integrated with an activated carbon bed to reduce radon concentration levels in homes. The design of such an integrated unit requires equilibrium isotherms and breakthrough curves of radon on these adsorbents. The radon adsorption on activated carbon, silica gel (grade 40), and molecular sieve-13X was measured at 288, 298, and 308 K both under static and dynamic conditions. The applicability of these data to the design of a radon-removal unit that could operate in conjunction with a dehumidification process is discussed.

INTRODUCTION

The health risks from exposure to radon daughters have become a major national concern, with the incidence of lung cancer from the inhalation of radon daughters currently recognized as being second to cigarette smoking. In a recent report, the Environmental Protection Agency (EPA) estimated that 16,000 radon-induced lung cancer deaths may occur annually in the U.S. Therefore, the development of effective methods for reducing radon concentrations in indoor air is of great interest. Increased ventilation, subslab ventilation, and the removal of soil gas are some of the methods that are currently employed to reduce indoor radon levels, although the effectiveness of increased ventilation depends on outdoor radon concentrations. Recently, it was reported (Barron 1991) that outdoor radon concentrations in a number of Canadian cities are higher than those indoors, with similar results being expected in such U.S. border states as North Dakota and Minnesota, where increased ventilation may not be an effective removal method. An alternative method may be to adsorb radon on solid adsorbents such as activated carbon and solid desiccants. Although silica gel and molecular sieves have been used extensively in solid-desiccant-based dehumidification systems, their ability to remove radon is not well documented. In order to make such processes attractive alternatives to conventional air-conditioning systems, the removal capabilities of solid adsorbents for radon and other indoor air pollutants must be understood and improved. Solid adsorbents interact with radon by the adsorption process. Since the amount of radon that can be adsorbed on solid adsorbents depends on temperature and its gas-phase concentration—and radon concentration levels can vary widely from residence to residence—radon equilibrium data (adsorption isotherms) are, therefore, required at different temperatures. Adsorption data for radon on activated carbon, silica gel, and molecular sieve-5A were reported between 1940 and 1960 and these data suggest that activated carbons have better radon adsorption and retention capabilities than both silica gel and molecular sieves. However, radon adsorption data are not given on commercially available silica gel and molecular sieve-13X at the present time. Because these two desiccants are frequently used in desiccant-based air-conditioning systems, any incidental removal of radon by these adsorbents is of considerable value. Note that molecular sieve-13X, which has a large pore diameter, is expected to have a higher adsorption capacity than either the 4A or 5A investigated earlier by Coleman et al. (1957).

The mechanism of radon and water vapor co-adsorption on solid adsorbents is not properly addressed in the literature. If activated carbon beds are used directly, they are expected to operate under conditions ranging from 20% to 80% relative humidity. Therefore, water vapor will play an important role in any process designed to remove radon. The literature on radon adsorption suggests that the presence of water vapor significantly reduces the radon-adsorption capacity of carbon. Therefore, an integrated system in which water vapor is removed first by a section packed with a desiccant material, followed by

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an activated carbon bed to remove radon, may be a better approach than using a carbon bed alone. Some diffusion barrier carbon canisters, which are used to measure radon concentrations in homes, also employ a small bag of silica gel in the gas-diffusing path to prevent water vapor adsorption by carbon. Because the diffusion of radon through silica gel will depend upon its adsorption capacity, adsorption isotherms of radon on silica gel and molecular sieve are also necessary for the proper design of diffusion barriers. The objective of this research is to conduct a comprehensive experimental study of radon adsorption on activated carbon and solid desiccants by both static and dynamic methods. In the static method, the equilibrium isotherm data of radon were obtained on three types of commercially available adsorbents: a coal-based activated carbon, silica gel (grade 40), and molecular sieve-13X. The radon concentrations in the gas and solid phases were measured simultaneously at equilibrium. In the dynamic method, experimental breakthrough curves of radon from the dry and moist nitrogen streams were measured at three temperatures. The applicability of these data to design a radon-removal process that would operate in conjunction with a dehumidification system is discussed.

TABLE 1
Properties of Solid Adsorbents

Property	Activated Carbon	Silica Gel (Grade 40)	Molecular Sieve 13X
Particle Size, Mesh[b]	6 × 16	6 × 12	8 × 12
Surface Area, S (m^2/g)[a]			
Micropores	823	663	294
Meso and Macropores	50	9	101
Total	874	672	395
Pore Volume, V (cm^3/g)[a]			
Micropores	0.47	0.38	0.14
Meso and Macropores	0.10	0.02	0.27
Total	0.57	0.40	0.41
Average Pore Diameter, 4 V/S (Å)[a]	26	24	10[c]
Bulk Density (g/cm^3)[b]	0.60	0.72	0.72
Moisture Content as Shipped (% wt.)[b]	<1	—	<1.5

[a]Analysis made by Porous Materials, Inc., Ithaca, NY.
[b]Analysis provided by the manufacturer.
[c]Based on total surface area and pore volume.

MATERIALS AND EXPERIMENTAL PROCEDURE

Materials

Properties of the adsorbents used in this study are given in Table 1. Radon gas was generated from a flowthrough source by flowing dry nitrogen gas from a cylinder. The radon source is a sealed container containing dry powder of ^{226}Ra with a stated activity of 22.6 kBq that is capable of producing radon gas at a constant rate of 2.847 Bq/min (76.87 pCi/min). The radon concentration was measured by a Lucas cell along with an AB-5 radiation monitor. The Lucas cell, which is coated inside with silver-activated zinc sulfide, has a very high scintillation efficiency and is used primarily for detecting alpha particles. When alpha particles strike the scintillator, extremely weak light output pulses of the scintillator are converted into electrical signals that are registered by a counting circuit.

Static Measurements

The adsorption isotherm data were obtained using an all-glass apparatus. The radon concentrations in the solid and gas phases were measured simultaneously once radon reached radioactive equilibrium with its daughter products and the adsorption equilibrium between the gas- and solid-phase concentrations was attained. In the solid phase, counts that were due to gamma activity of the ^{214}Pb and ^{214}Bi were recorded by an NaI(Tl) detector, whereas in the gas phase, counts based on the decay of alpha-emitting radon daughters (^{218}Po and ^{214}Po) were obtained from the Lucas cell. Standard procedures were followed to obtain the counts from the NaI(Tl) detector and the Lucas cell; interested readers may refer to Knoll (1979) for a detailed explanation of the subject. The details of the experimental apparatus and procedure, the methods of calibration prior to each run, and the radioactivity calculation are described by Hassan et al. (1991). The error in the count rate of the gas phase was determined at the one-sigma significance level and it ranged from 4.79% to 11.92%. The minimum detectable activity due to background count was found to be 21.2 counts per minute at the three-sigma significance level.

Dynamic Measurements

A glass column 4 cm in diameter and 36 cm long was packed with approximately 50 grams of the adsorbent sample so that a bed-diameter-to-particle ratio of 17.5 could be maintained in the bed to avoid channeling and wall effects. A superficial gas velocity of 0.198 L/min^{-1} was maintained in the bed during experimental runs. The concentration of radon in the inlet nitrogen stream was fixed at 220 ±20 pCi/L throughout the experiments, except when the effect of flow rate was investigated. The adsorbent was regenerated by flowing dry nitrogen at 473 ±0.1 K through the bed for 10 hours. After regeneration, the adsorbent bed was cooled to the desired adsorption temperature. The background counts due to atmospheric radioactivity, which always varied between 10 and 12 counts per minute, were measured using a Lucas flow-

through cell along with an AB-5 radiation monitor. Nitrogen was next directed to the radon source, which was flushed for one hour at a constant flow rate before it was introduced into the column. The radon concentration at the bed outlet was monitored at five-minute intervals by using the Lucas cell until breakthrough was complete and the counts of the effluent reached a constant value. Following the adsorption run, the regeneration of the bed was continued by flowing radon-free nitrogen through the bed. This was continued until the count rate of the column effluent was equal to the background count of the Lucas cell.

RESULTS AND DISCUSSION

The equilibrium isotherm data for radon on activated carbon, silica gel (grade 40), and molecular sieve-13X at 298 K are shown in Figure 1. The uptake of radon by activated carbon was considerably higher than that of silica gel and molecular sieve-13X, with silica gel exhibiting a relatively low radon adsorption capacity. However, molecular sieve-13X exhibited a higher affinity for radon than was found for Type 5A (Coleman et al. 1957), which may be due in part to the large pore diameter of 13X. All data exhibited a Type III isotherm, in conformity with the literature (Przytycka 1961; Burtt and Kurbatov 1948; Coleman et al. 1957).

Experimental breakthrough curves for radon on activated carbon, silica gel, and molecular sieve-13X were measured at 288, 298, and 308 K using bed lengths for activated carbon, silica gel, and molecular sieve of 10.16, 11.43, and 13.97 cm, respectively. It should be noted that the same amount of adsorbent (50 g) was used in each case and the difference in bed length was due to different bulk densities of the materials. The flow rate of nitrogen through the radon source was maintained at 0.35 L/min for all experimental runs and produced a steady inlet radon concentration of approximately 220 pCi/L (radon partial pressure of 1.2×10^{-12} mmHg). The total pressure drop across the beds ranged from 10 to 15 mmHg. Breakthrough curves for these three systems are shown in Figures 2 through 4. As can be seen from these figures, the steady-state outlet concentration of radon was lower than the inlet concentration.

According to Madey et al. (1978), the steady-state concentration of radioactive gas at the outlet of a column

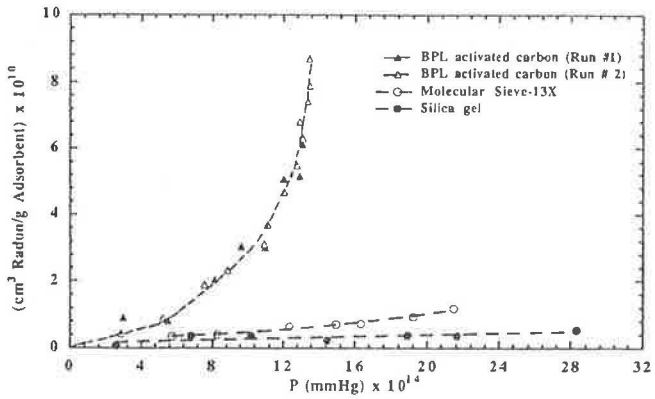

Figure 1 *Adsorption isotherms of radon on various solid adsorbents at 298 K.*

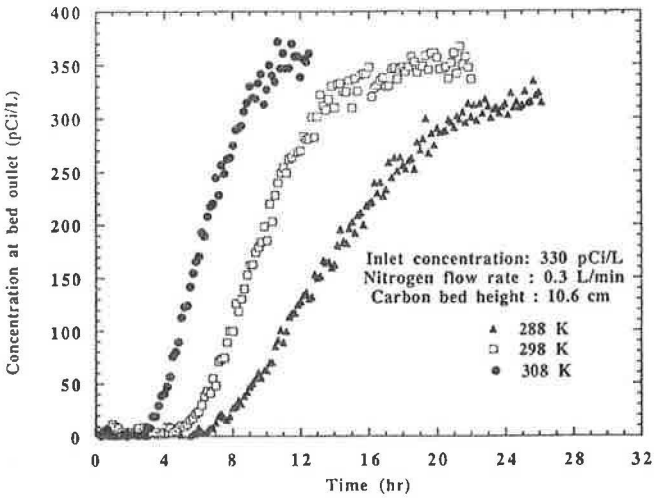

Figure 2 *Experimental breakthrough curves for radon adsorption on activated carbon.*

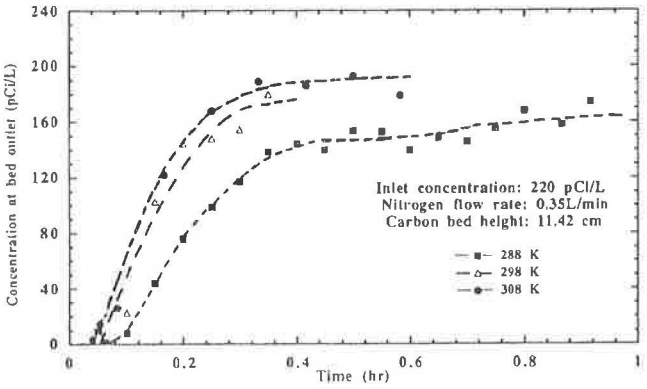

Figure 3 *Experimental breakthrough curves for radon on silica gel.*

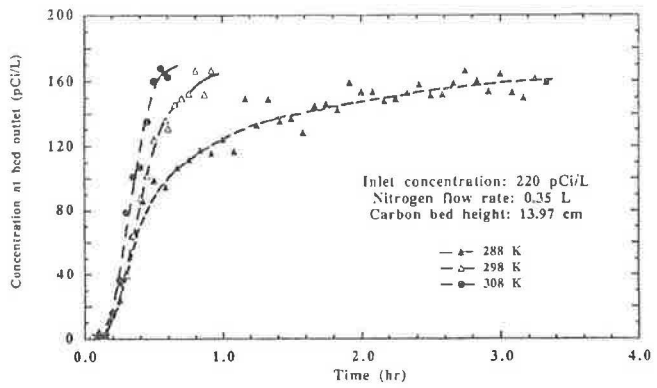

Figure 4 *Experimental breakthrough curves for radon on molecular sieve 13X.*

of length L is related to the inlet concentration through the expression

$$C_{out} = C_{in} \exp(-\gamma L) \quad (1)$$

where

$$\gamma = \frac{u}{2D}[(1 + 4DB\lambda/u^2)^{1/2} - 1], \quad (2)$$

λ is the decay constant for the radioactive gas, u is the superficial gas velocity (cm/s), B is the effective adsorptivity (dimensionless), and D is the effective dispersivity (cm^2/s). To verify this phenomenon, a separate experiment was carried out in which the radon concentration at the inlet was monitored continuously along with the outlet concentration for 48 hours. It was found that the count rate at the outlet of the column reached a steady-state value that was lower than the inlet value. It was also noted that the various solid adsorbents led to different steady-state radon concentrations at the column outlet for the same inlet concentration. As the bed temperature was increased, the shape of the breakthrough curves became steeper and radon broke through the column at a faster rate. However, the uptake of radon by the silica gel did not change significantly with the temperature. A similar result was observed for radon adsorption on molecular sieve-13X. A comparison of the characteristic breakthrough curves of radon on the three adsorbents (carbon, silica gel, and molecular sieve-13X) at 298 K is shown in Figure 5. This result indicates that radon adsorption and retention by silica gel and molecular sieve-13X beds are considerably lower than that of activated carbon. It should be noted that a state of dynamic equilibrium was established due to the decay of radon even in the silica gel and molecular sieve beds. Therefore, a small amount of radon will be removed in any desiccant bed on a continuous basis and can be considered a plus during the dehumidification process. The effects of repeated adsorption/regeneration cycles for radon on activated carbon are shown in Figure 6. The adsorbent was regenerated after each cycle by passing radon-free nitrogen at a temperature of 200°C through the bed for 12 hours. It can

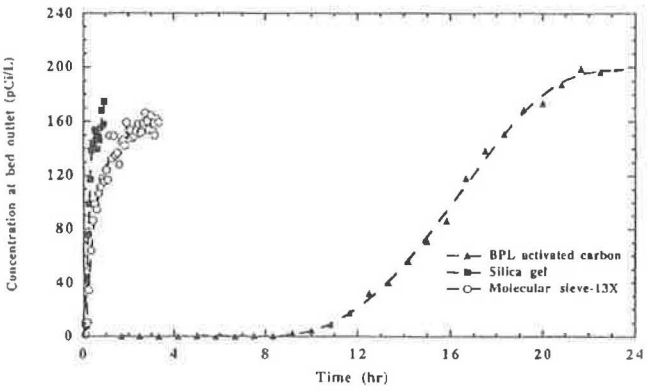

Figure 5 Radon breakthrough curves on various solid adsorbents at 288 K.

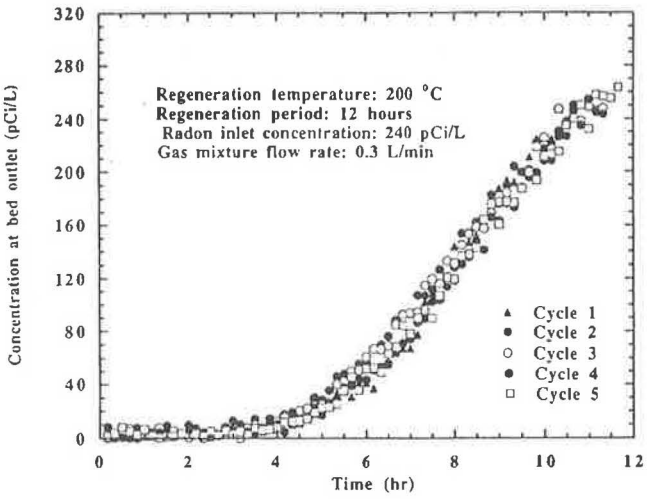

Figure 6 Adsorption/regeneration cycles of radon on activated carbon at 298 K.

be seen from the figure that after five cycles the bed would adsorb the same amount of radon as the fresh sample. Radon is adsorbed as a gas into the pores of activated carbon and is held there by various adsorption forces. Radon further decays into ^{214}Pb and ^{214}Bi (called radon daughters), which are in the solid phase and remain deposited in the pores. Therefore, a carbon bed can not only adsorb (capture) radon from the air, it can also retain the daughter products in the bed. However, it is interesting to note that these daughter products do not reduce the availability of active sites, as found from repeated adsorption/regeneration cycles. The adsorption capacity of the bed did not change significantly during five cycles.

The breakthrough curves of radon from radon and water vapor mixtures on activated carbon were measured at different relative humidities and are shown in Figure 7. The steady-state radon concentration at the column outlet was found experimentally to be greater than the inlet concentration. This may be due to increased radon plate-out on the inside of the Lucas cell. A similar observation was reported earlier by Boncanegra and Hopke (1988), who found that the presence of water vapor caused

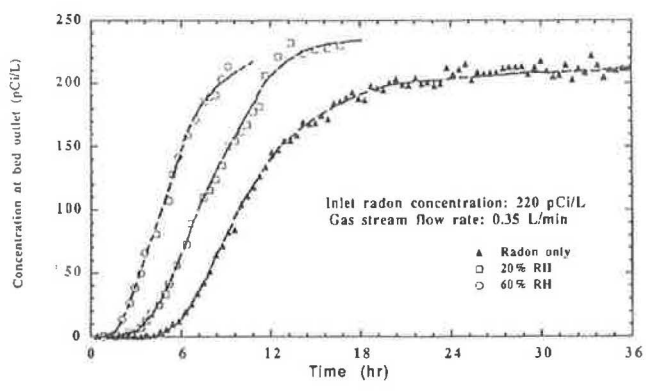

Figure 7 Breakthrough curves for radon from moist nitrogen stream on activated carbon at 298 K.

neutralization of ^{218}Po ions and resulted in their deposition in the detector. The adsorption of radon on activated carbon from humid nitrogen is considerably less than that from a dry nitrogen stream. The presence of water vapor in the nitrogen stream reduced the uptake capacity for radon due to the strong competition by water vapor molecules for the available sites. However, it is interesting to note that radon broke through the bed at approximately the same time regardless of the relative humidity of the nitrogen stream. Water vapor molecules, having a stronger affinity for activated carbon than radon, might be adsorbed near the inlet section of the bed at a faster rate. As the adsorbed water front progresses through the bed, it displaces radon and results in a faster radon breakthrough from the bed.

CONCLUSIONS

The adsorption isotherm data of radon on silica gel, molecular sieve 13X, and carbon suggest that carbon is the better adsorbent for radon. The adsorption capacity of silica gel and molecular sieve is negligible when compared to carbon. Equilibrium isotherms of radon, such as the ones shown above, can be used for measuring radon concentrations in homes, provided accurate isotherm data are available for the activated carbon. From knowledge of the solid-phase radon concentration, the equilibrium isotherms corresponding to the particular field conditions, temperature, and humidity can be used to determine the concentration of radon in the gas phase. Another application for the dynamic and static adsorption data is in the design of an adsorber unit for removing radon from indoor air. Since the radon-adsorption capacity of carbon can decrease significantly in the presence of moisture, the design of such a unit may range from a single packed column containing an activated carbon section downstream of the desiccant bed to a complex system of multiple columns.

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